Attractability and Separability of Hematite in High Gradient Magnetic Separation(ヘマタイトの高勾磁選における捕捉性と選別性)

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論　文　内　容　要　旨

Abstract

High gradient magnetic separation technology has a considerable significance for many resource utilization and pollution control problems facing the world to day. These separators are a new departure which extends the application of magnetic separation technology to mineral system with a wide range of magnetic properties, including both strongly magnetic and very weakly magnetic minerals. High gradient magnetic separation has extended the use of magnetic separation to even weakly paramagnetic particles with diameters of the order of a micron.

Until comparatively recently, high intensity separation has been confined solely to dry ore and has been used commercially since 1908. Field intensity over 1.6×10^{-6} [A/m] can be obtained in some high intensity machines. Within the last few years high gradient magnetic separators have been developed, that allow weakly paramagnetic colloidal particles to be extracted from a fluid moving through the separator.

The high gradient magnetic separators cosist of ferromagnetic wires, stainless steel wool ……… etc., occupying about 5−10% of the space magnetized by a uniform magnetic field, so that magnetic field gradients as high as 8.0×10^{7} [A/m]/m can

be achieved. In addition, high gradient magnetic separation is an important process not only for the beneficiation of iron ores, but also in the treatment of paramagnatic nonferrous minerals and nonmetallic minerals.

The matrix is held in a canister into which the slurry is fed. As the slurry passes through the matrix, the magnetic particles are captured at the magnetized fibers while some other particles are mechanically captured. Periodically, the magnetic field can be removed and the matrix is flushed with water to remove the captured materials.

Briefly we can summerize the applications and uses of high gradient magnetic separation as follows :

1. Purification and enrichment of materials in liquid suspensions (for example, removing the micron size of iron content from clay in clay industry).
2. Beneficiation of so called nonmagnetic taconite ore as well as ore of uranium, molybdenum and many of transition metal elements.
3. Coal desulpherization and deashing as well as water purification.
4. Separation and concentration of hematite.
5. Separation of sulphide minerals.
6. Water treatment.

In addition, we can summerize the promising application as follows :

1. Decinderization of coal.
2. Separation of rare earth minerals.
3. Treatment of heavy oils.
4. Treatment of waste water from various industries and mines.
5. Treatment of restoring water in boilers at steam and atomic plants.
6. Treatment of oil containing waste water and sludges.
7. Elimination of iron bearing materials from atmospheric air.
8. Elimination of arsenic and silica from hydrothermal solutions.
9. Application to food and drug industries.
10. Elimination of phosphated materials, suspended solids, algae and bacterium coli from livelihood waste water and river dranage.
11. Recovery of uranium from sea.

The degree of particle attractability and separability depends on many operational parameters, such as magnetic field intensity, pulp density, pulp flow rate, particle size, feed grade, diameter of matrix element, capture time, packing density, susceptibility of treated magnetic particle, pulp viscosity.........etc.

Theoretical treatments of capture process on a fiber and the collection parameters for an assembly of wires have been given by many investigators. As well as, there are many experimental data on the performance of high gradient magnetic separation system. However, no quantitative data on the collection process neither comparative results with the conventional separatos are available yet.

Various researches have been conducted on the capture process in order to study the shape and the phenomenon of buildup using visible systems of instrumentation. The previous studies were mainly concerned with a single magnetic particle. On the other hand, there is a lack of researches on the separation of one mineral from another by using high gradient magnetic separation. As well as, the effect of different operational parameters on both the attractability and separability of magnetic mineral mixed with nonmagnetic one did not receive much attention from the experimental point of view. In addition, there is just a few researches on the locking effect phenomenon and no comparative studies between high gradient magnetic separators and conventinal magnetic one are available.

We have discussed in detail, the theories of high gradient magnetic separation which had been reviewed by different authors.

This study is mainly concerned with the attractability and separability of hematite at different operational parameters, from mixtures of hematite and quartz by using a special design of fine stainless steel wires. The wires are arranged in a special layout,to form the matrix unit, and inserted inside the canister such that the plane of matrix and the direction of the slurry flow are parallel and both are perpendicular to the direction of the magnetic field. This configuration is known as what is called axial configuration or case C, and we used it in all experimental work.

Six operational parameters were selected for this study. These parameters are wire diameter, D_w, particle size, D_p, feed grade, f, flow rate, u, pulp density, p, and magnetic flux density, B. This study have been carried out on the Brazilian hematite, the Egyptian hematite, and the Egyptian pyrolusite as a magnetic components, while the Japanese quartz was selected as a nonmagnetic one.

All experiments were carried out on a mixed samples of magnetic and nonmagnetic powders suspended in distilled water. The effect of each of the above parameters on the attractability and separability was investigated each at a time, while the other variables were kept constant.

Experimental apparatus

Experimental set-up:

The layout and instrumentation of the installation which we have used in this study comprises from the following units:

1. A 3 liter feed tank, conical in shape and fitted at the bottom with a glass valve in order to control the flow.
2. A stirrer driven at 2200 r.p.m. is fitted at the center of the feed tank in order to prevent the settling of solids and to ensure efficient mixing.
3. An electromagnet with a variable gap. The magnetic field reaches 7 KG when the gap is 76 mm. The electromagnet is equipped with circular pole pieces 70 mm in diameter.
4. A power supply, powered the magnet and it has a maximum current output of 30 A and a maximum voltage output of 60 volts.
5. A flowmeter, graduated between 1 and 6 l/hr is installed on the discharge line in order to measure the flow rate of the slurry.

High gradient magnetic separation unit:

High gradient magnetic separation unit consists of two parts:

1. Matrix unit: it is an array of 24 fine expanded stainless steel wires(ss. USU 430) each 1.0 mm in diameter, and 90 mm in length. The space between the fine expanded wires was kept at 5.0 mm by a special design of copper grate. Another three matrix units were designed similar to the former one with three different wire diameters, namely, 0.8, 1.5, and 2.0 mm.
2. Canister: it made of transparent acrylic pipe 5.5 mm in thickness, 40 mm inner diameter, 51.0 mm outer diameter, and 1300 mm in length.

Properties of sample

Three magnetic powders were investigated, which are the Brazilian hematite, the Egyptian hematite, and the Egyptian pyrolusite, while the Japanese quartz was used as a nonmagnetic powder.

Firstly, our study was conducted, mainly, on the Brazilian hematite and the Japanese quartz. A representative sample from the original head sample, which supplied by NIHON KŌKAN Co. and yielded from Companhia Vale do Rio Doce, was collected by the technique of coning and quartering, and then the tested size fractions have been obtained by conducting a pulverising tests(i.e. crushing by jaw crusher and cone crusher, and then grinding by ball mill) as well as sizing tests (by Ro-tap testing seiving screens). The tested size fractions have been selected to be −100+150, −150+170, −170+200, −200+250, −250+270, −270+325, −325+400 mesh. This technique of sample preparation have been used for all tested powders.

A representative sample from the pulverised product of all tested powders were chemically analysed. The results of the chemical analysis showed that the Brazilian hematite contains 91.96, 0.96, 0.07, 0.48, 0.05, 0.043, 0.10, 0.022, and 0.004% of Fe_2O_3, SiO_2, CaO, Al_2O_3, MgO, TiO_2, MnO, P, and S respectively. On the other hand the Egyptian hematite was found to contain 91.27, 0.84, 0.14, 0.18, 0.13, 0.014, 1.36. 0.195, and 0.203% of the same elements respectively. With respect to the Egyptian pyrolusite the percentage of MnO_2 was found to be 85.73%, while the percentage of SiO_2 in the Japanese quartz was found to be 98.0%.

In addition, X–ray powder diffraction analysis of collected samples were carried out. The X–ray powder diffraction patterns of the investigated specimens were obtained by Rigaku–Denki X–ray diffractometer. The results of X–ray diffraction analysis showed that the Brazilian hematite is mainly hematite while the Egyptian hematite contains admixed goethite. The X–ray analysis of the Egyptian pyrolusite showed that the sample is β-MnO_2.

In our study the magnetic properties of both the magnetic powder, which includes the Brazilian hematite, the Egyptian hematite, and the Egyptian pyrolusite and the stainless steel wire of the matrix unit were investigated by using magnetic balance.

In addition, the flow conditions were investigated and we have realized that all particles of all tested powders settle within laminar flow conditions. Also, density measurements for all tested powder have been done. The results showed that the densities of the Brazilian hematite, the Egyptian hematite, the Egyptian pyrolusite, and the Japanese quartz were 5.23, 3.96, 4.44, 2.62 g/cm^3 respectively.

Experimental procedure

The experimental procedure can be summerized as follows:

1. The acrylic pipe as well as the feed tank are filled with deionized water such that the feed tank contains the required volume.
2. The required suspention is prepared by adding the weighed amount of magnetic powder and quartz, with desired ratio for a given test, into the mixing feed tank. Sodium silicate solution was used to adjust pH at 10 in order to prevent the heterogenous coagulation. The solids are suspended in the feed tank while the stirrer is working. Five minutes is used to obtain efficient mixing.
3. The magnetic field is firstly established at the desired value, while stirring the slurry in the feed tank. The valve of the feed tank is then opened gradually such that the slurry moves at gentle rate into the canister (i.e. acrylic pipe), and then through the matrix unit which is located in the magnetic field between the two poles of the magnet. The rate of the flow is regulated by means of flowmeter's valve.

4. After the saturation time, the valve of the feed tank is closed and the canister is emptied at the same flow rate, then the magnetic field is turned off. The lower part of the canister is detached and the matrix unit is carefully lowered into a plastic container of almost similar dimensions. The captured magnetic materials on the wire of the matrix unit as well as on the wall of the canister are washed off, individually, gently and throughly with distilled water.
5. Both captured and uncaptured materials are subjected to gravimetric analysis. In all experiments, the total captured weight, on the wire and wall, magnetic captured weight, nonmagnetic capture weight, magnetics recovery, nonmagnetics recovery, and ratio of enrichment are calculated.

   The recovery is calculated according to the following equation:

$$R = \frac{Cc}{Ff} \times 100$$

where,

R : recovery

C : weight of concentrate ,[kg]

c : assey of concentrate

F : weight of feed, [kg]

f : assey of feed

while the ratio of enrichment can be calculated according to the following equation:

$$\varepsilon = \frac{f_c}{f_f}$$

where,

ε : ratio of eurichment

f_c: grade of concentrate

f_f: grade of feed

The studied parameters were as follows:

1. magnetic flux density, B,[T]
2. flow rate, u, [m/s]
3. particle size, D_p , [m]
4. wire diameter, D_w , [m]
5. pulp density, p ,
6. percentage of the magnetic materials of feed, f ,
7. kind of treated powders

The effect of each of the above parameters on the attractability and separability was investigated, each at a time, while the other variables were kept constant.

Experimental Results

Attractability of hematite

In order to study the attractability of hematite by high gradient magnetic separation firstly, we investigated the variation of the capture amount with the experimental time under different operational conditions using two kinds of hematite ores, namely, the Brazilian hematite, and the Egyptian hematite as well as the Egyptian pyrolusite which was used as application for another magnetic material. Some parameters have been chosen under consideration, that is, feed grade, slurry flow rate, pulp density, particle size of feed material, magnetic flux density in vacant canister, and diameter of wire composing the matrix unit.

These parameters were changed in different ways and the capture amount as well as the total capture amount were measured at various intervals of experimental time. The total capture amount was expressed as the total weight of both particles attracted on the wires of the matrix unit and those captured on the wall of the acrylic pipe(canister).

It has been concluded from these experiments that the total capture amount increases with the capture time, and then it reaches to saturation state at capture time equals to 30 minutes. Also we have concluded that the higher the flow rate is the lower the total capture amount is.

The relation between the total capture amount and the capture time was found to be as in the following equation:

$$C_t = C_{t\infty} \ (1 - e^{-kt})$$

where,

C_t = total capture amount, [kg]

$C_{t\infty}$= total capture amount at saturation,[kg]

t = capture time, [s]

k = velocity coefficient , [s^{-1}]

By using the least squares method we could obtained the following empirical equation:

$$C_t=(-0.141u + 7.060 \times 10^{-4})[1-e^{-(0.0675u + 1.535 \times 10^{-3})}] \pm 1.07 \times 10^{-5}$$

where, u is the flow rate in m/s.

Another parameter which is the feed grade, f, have been investigated under the same previous operational conditions and we have concluded that the total capture amount increases gradually with capture time till saturation is obtained at 30 minutes. As well as, the higer the value of feed grade is the higher the value of total capture amount is. It was also evident from the relation between the total capture amount and the capture time, that the relation follows the same previous equation, namely, $C_t = C_{t\infty}(1-e^{-kt})$.

The following empirical equation have been developed using the least squares method:

$$C_t=[8.508\times10^{-4}(f/100)+4.831\times10^{-5}][1-e^{-[-4.190\times10^{-5}(f/100)+1.937\times10^{-3}]t}]$$

$$\pm 1.82\times10^{-5}$$

where, f, is the feed grade.

The total capture amount depends on a various properties, therefore, the dependence of the capture amount on these various parameters was investigated mathematically by using dimensional analysis based on π theory. The follwing two equations have been developed for the Brazilian hematite, and the Egyptian hematite respectively.

$$C_{t\infty}= 8.46\times10^{-6}\rho_a\cdot D_p^{0.20}\cdot S^{0.15}\cdot \chi_m^{-0.86}\cdot u^{-0.32}\cdot f^{0.78}$$

$$C_{t\infty}= 2.67\times10^{-2}\rho_a\cdot D_p^{1.55}\cdot S^{0.15}\cdot \chi_m^{0.78}\cdot u^{-0.61}\cdot f^{0.63}$$

where,

ρ_a : apparent density of pulp, [kg/m³]

S : total capture area of the magnetic wires, [m²]

χ_m : magnetic susceptibility of hematite, [H/m]

By using the previous equations we calculated the total capture amount, $C_{t\infty_{cal.}}$, and these values were compared with the values of the total capture amount which we have got experimentally, $C_{t\infty_{exp.}}$. Excellent coincidence was obtained in this comparison and the relation between calculated **and** measured values was found to be of a straight line nature.

In addition, it was found that the most effective parameters on the total capture amount are magnetic susceptibility of the Brazilian hematite, χ_m, and the feed grade, f, while in the case of the Egyptian hematite the most effective parameters were found to be the magnetic susceptibility, χ_m, and the particle size, D_p.

The relation between total capture amount, $C_{t\infty}$, and the wire capture amount, C_w, for all tested powders have been investigated, and the relations were found to be a straight line relationships. The equations which show these relations can be expressed as follows:

$C_w = 0.905\ C_{t\infty} - 53.48\times10^{-6} \pm 2.62\times10^{-6}$ (Brazilian hematite)

$C_w = 0.906\ C_{t\infty} - 32.04\times10^{-6} \pm 3.60\times10^{-6}$ (Egyptian hematite)

$C_w = 0.962\ C_{t\infty} - 22.77\times10^{-6} \pm 1.51\times10^{-6}$ (Egyptian pyrolusite)

We have expressed what is called critical capture time, t_c, by empirical formula, i.e.

$$t_c = \frac{0.90\ C_w\ \varepsilon}{Q\ \rho_a\ (p/100)\ \alpha}$$

where,

C_w : the amount captured on the magnetic wire at saturation, [kg]

ε : ratio of enrichment

Q : volumetric flow rate of the slurry, [m³/s]

ρ_a : apparent density of the pulp, [kg/m³]

α : correction factor (to correct the effect of particles sedimentation on the top of the grate of the matrix unit)

The critical capture time is a very important parameter in high gradient magnetic separation, hence, we have calculated the critical capture time using the previous equation and compared the calculated values, t_{cc}, with the measured values, t_{cm}, and we have realized that excellent coincidence could be obtained.

The variation of capture amount from the starting time till saturation state was found to be of a transitional phenomenon, this transitional state was judged by the value of the velocity coefficient, k, which we have explained earlier. It was found that the bigger the value k is the sooner the capture amount reaches to the saturation value.

We have been surveyed how the value k varies according to the different operational parameters, and by using the same technique of dimensional analysis we have investigated the dependence of k on the various parameters in the cace of the Brazirian hematite and the Egyptian hematite. The following equations have been developed respectively:

$$k = 6.43 \times 10^{-21} S^{0.65} \cdot D_p^{-3.00} \cdot \chi_m^{3.00} \cdot u^{-0.10} \cdot f^{-0.38}$$

$$k = 1.56 \times 10^{-25} D_p^{-4.00} \cdot \chi_m^{0.70} \cdot u^{-1.70} \cdot f^{-0.30}$$

These results showed also that the particle size, D_p, and the magnetic susceptibility, χ_m, influence much on k in the cace of the Brazirian hematite while D_p and the flow rate, u, inflnence on the value of k for the Egyptian hematite.

Separability of hematite

The relation between ratio of enrichment, ε, and the percentage of magnetic materials in feed, f, have been investigated for all tested powders using high gradient magnetic separation.

Our results were compared with the results which have been obtained previously by Yashima using conventional magnetic separators and highly magnetic powders. It was evident from the comparison that, treating highly magnetic powder (i.e. magnetite and iron sand) by conventional magnetic separators (i.e. drum type and Crockett type) gives the same result as that obtained by treating weakly magnetic particle (hematite) in high gradient magnetic separators.

In addition, the effect of different operational parameters on the ratio of enrichment have been investigated and we concluded that only the feed grade affects the ratio of enrichment while all the other parameters do not affect. The relations between ratio of enrichment, ε, and the feed grade, f, for all tested powders were found to be as follows:

$\varepsilon = (\frac{f}{100})^{-0.895}$ ± 0.012 (Brazilian hematite)

$\varepsilon = (\frac{f}{100})^{-0.903}$ ± 0.038 (Egyptian hematite)

$\varepsilon = (\frac{f}{100})^{-0.904}$ ± 0.017 (Egyptian pyrolusite)

By investigating the relation between ε and f for all tested parameters, we have obtained exactly the same separability curve as we obtained for only one parameter and the relations were found as follows:

$\varepsilon = (\frac{f}{100})^{-0.895}$ ± 0.012 (Brazilian hematite)

$\varepsilon = (\frac{f}{100})^{-0.940}$ ± 0.380 (Egyptian hematite)

$\varepsilon = (\frac{f}{100})^{-0.904}$ ± 0.017 (Egyptian pyrolusite)

Repitition technique as one of the most advantageous way to get a concentrate of high grade has been tried with all tested powders and we have got a concentrate of, more or less, 100.0% grade after repeating the separation process for 4 times (for feed grade 10.0%) and after repeating the separation process for 2 times (for feed grade 80.0%). The same results have been obtained for all tested powders, namely, the Brazilian hematite, the Egyptian hematite, and the Egyptian pyrolusite.

All experiments which we have done were carried out by using the experimental unit of high gradient magnetic separation. Therefore, it was interesting to compare the attractability and separability of hematite and pyrolusite which we have obtained with that of the actual high gradient magnetic separator (SALA HGMS).

Three different kinds of matrixes have been selected to conduct this comparison, namely, coarse stainless steel wool, fine stainless steel wool, and mesh matrix type. The operational parameters were kept constant in all cases, and more or less the same as that of the experimental unit. The separability curves have been investigated and the relation between ε and f was studied.

It was found that, in the case of coarse stainless steel wool, the separability curve is the same for all tested powders and it was almost the same as in the case of our experimental unit. On the other hand, for mesh type and fine stainless steel wool the value of the exponent α was found smaller. It can be attributed to the effect of mechanical capture as well as packing density. The results of this study are clear in the following equations:

$$\varepsilon=\left(\frac{f}{100}\right)^{-0.940} \pm 0.17$$ (Brazilian hematite, coarse stainless steel wool)

$$\varepsilon=\left(\frac{f}{100}\right)^{-0.925} \pm 0.17$$ (Egyptian hematite, coarse stainless steel wool)

$$\varepsilon=\left(\frac{f}{100}\right)^{-0.894} \pm 0.13$$ (Egyptian pyrolusite, coarse stainless steel wool)

$$\varepsilon=\left(\frac{f}{100}\right)^{-0.337} \pm 0.05$$ (Brazilian hematite, fine stainless steel wool)

$$\varepsilon=\left(\frac{f}{100}\right)^{-0.716} \pm 0.04$$ (Brazilian hematite, mesh matrix type)

The attractability of the magnetic powders have been investigated by using SALA HGMS, and the sam results as in the cace of the experimental unit were obtained.

Since the recovery is one of the most important parameters which describe the extent of the separation process, therefore, the effect of different operational parameters on the recovery of both hematite and quartz was studied.

All the previous experiments have been carried out by using closed size of both hematite and quartz. Another sets of experiments have been conducted on particles of different size distributions, and the results showed that better separation could be obtained when the particle size of quartz was much coarser than that of hematite. The results showed also the same tendency of separability curves.

In addition, some calculations for the magnetic forces affecting between wire and particle as well as between particle and particle have been done.

Also some studies on the characteristics of the magnetic field and field gradient have been conducted.

Conclusions:

The attractability and the separability of hematite in high gradient magnetic separation have been studied. The comparison between the experimental unit of high gradient magnetic separation and conventional magnetic separators as well as SALA high gradient magnetic separator, from separability and attractability stand point of view, has been reported.

The relations between total capture amount and the different operational parameters were formulated.

We have explained the transitional phenomenon of the capture process, and we judged it by what is called "velocity coefficient". The relation between the velocity coefficient and the operational parameters were formulated.

We have introduced an empirical formula for calculating the critical capture time.

A straight line relationship between wire capture amount and total capture amount were concluded.

The results which we have obtained for all tested powders were the same.

審 査 結 果 の 要 旨

高勾配磁選は微粒弱磁性鉱物の選別に有効な方法として近時注目されているが，その選別機構については従来磁界内における単一磁性粒子の挙動が明らかにされているのみで，非磁性粒子が共存する場合の選別機構は未解明の状態であった。本論文はヘマタイトと石英の混合試料を用いて高勾配磁選の捕捉性と選別性を究明し，その設計と操作の基礎となる研究成果をまとめたもので，全編7章より成る。

第1章は緒論であり，本研究の意義と目的を述べている。

第2章では既往における単一磁性粒子の捕捉性に関する研究結果を総合的に検討して，以降の研究の指針をえている。

第3章では試作した高勾配磁選装置につき磁界の強さとその勾配，磁性細線周域の磁界特性を調べて本装置が基礎研究に十分な性能を有していることを述べている。

第4章では2種類のヘマタイトおよび比較に用いたマンガン鉱ならびに石英の化学分析，X線解析からこれらの試料が十分な品位であることを確かめ，比重と磁性測定結果を述べると共に全測定を湿式，層流範囲で行った点にも言及している。第5章は実験方法である。

第6章では実験結果を述べている。捕捉性については捕捉量の経時変化から求めた飽和捕捉量 $C_{t\infty}$ と給鉱品位，粒径，パルプ流速，細線総表面積等の操作要因との関係を次元解析によって導き，$C_{t\infty}$ の90%に達する時間を限界捕捉時間と定義してこれと操作要因との関係を明らかにし，また $C_{t\infty}$ に達する過渡特性も検討している。この結果はサイクリック操作の周期決定に指針を与える重要な研究成果である。一方給鉱品位による富鉱比の変化で選別性を表すと，この特性曲線は試料の種類と操作要因に関係なくほぼ一定の傾向を示し，また粗粒強磁性鉱物を低磁界条件で選別する在来の磁選のそれとも良好に一致することを見いだし，高勾配磁選の選別性は本質的に在来の磁選のそれと同じと見なしうるという結果をえ，さらにSALAの実用試験機によってもこれを確かめている。これは著者によって初めて明らかにされた重要な成果である。

第7章は結論である。

以上要するに本論文は，ヘマタイトの高勾配磁選における捕捉性と選別性を究明して，その設計と操作の基礎を明らかにしたものであり，鉱物処理工学とこれに関連する工業に寄与するところが少なくない。

よって，本論文は工学博士の学位論文として合格と認める。